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STUDIES ON RADIATIVE COLLISIONAL AND ULTRAVIOLET LASERS

FINAL REPORT

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)  Significant accomplishments of this program include: the first microwave pumping of an excimer laser; the first use of pulsed hollow-cathode technology to produce substantial quantities of core-excited metastable atoms; the use of these metastable atoms to delineate a partial Grotrian diagram for core-excited Na; the proposal of the concept of quasi-metastable quartet levels and the experimental verification of their importance in the column I metals; and the proposal for short wavelength systems based on super Coster-Kronig transitions. - 2 to 12		

## I. INTRODUCTION

This program has supported theoretical and experimental studies in several areas of laser physics particularly applicable to the construction of XUV and soft x-ray lasers. The work during this three-year period has been exceedingly successful. Numerous ideas and several experimental successes have resulted. Section II summarizes the key results of this program; Section III gives the list of publications; Section IV lists oral disclosures; Section V lists the personnel; and Section VI gives abstracts of all publications supported by this program.

We note that this program was undertaken jointly with the U.S. Army Research Office.

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## II. SIGNIFICANT ACCOMPLISHMENTS

The significant accomplishments of this program are as follows:

(1) Following our first demonstration of the microwave pumping of an excimer laser, such a laser was mode-locked for the first time.

(2) D. E. Holmgren, et al. used pulsed hollow-cathode technology to accomplish the first measurement of metastable quartet population of core-excited alkali atoms. These measurements showed that levels such as these which lie above a continuum retain their integrity even in the presence of high electron density.

(3) In a key theoretical publication the concept of quasi-metastable quartet levels was proposed. As shown by further experiment, these levels have radiative times on scale with their autoionizing times and are excellent candidates for XUV lasers.

(4) The first experimental demonstration of quasi-metastability was obtained by Holmgren, et al., who showed radiation at  $1091 \text{ \AA}$  in neutral Cs on scale with that of the strongest ion lines.

(5) Using the lowest metastable level of Na and laser transfer techniques a Grotrian diagram for the core-excited quartet manifold of Na I was defined. This was the first demonstration of such a technique in the extreme ultraviolet spectral region.

(6) In another key result we proposed the idea of selectively autoionizing laser systems based on super Coster-Kronig transitions.

(7) The work on Cs was confirmed in an experiment by Pedrotti and Dimiduk where harmonic generation at  $1091 \text{ \AA}$  was used to confirm the nature of the  $1091 \text{ \AA}$  transition.

(8) Using high-power pulsed microwave technology the quasi-metastability concept was shown to extend throughout column I.

### III. LIST OF PUBLICATIONS

1. S. E. Harris, J. F. Young, R. G. Caro, R. W. Falcone, D. E. Holmgren, D. J. Walker, J. C. Wang, Joshua E. Rothenberg, and J. R. Willison, "Laser Techniques for Extreme Ultraviolet Spectroscopy," in Laser Spectroscopy VI, H. P. Weber and W. Luthy, eds. (New York: Springer-Verlag, 1983), pp. 376-381.
2. P. J. K. Wisoff and J. F. Young, "Active Mode-Locking of a Microwave-Pumped XeCl Laser," IEEE J. Quant. Elect. QE-20, 195-197 (March 1984).
3. D. E. Holmgren, R. W. Falcone, D. J. Walker, and S. E. Harris, "Measurement of Lithium and Sodium Metastable Quartet Atoms in a Hollow-Cathode Discharge," Opt. Lett. 9, 85-87 (March 1984).
4. S. E. Harris, D. J. Walker, R. G. Caro, A. J. Mendelsohn, and R. D. Cowan, "Quasi-Metastable Quartet Levels in Alkali-Like Atoms and Ions," Opt. Lett. 9, 168-170 (May 1984).
5. D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris, "Grotrian Diagram of the Quartet System of Na I," in Laser Techniques in the Extreme Ultraviolet, S. E. Harris and T. B. Lucatorto, eds. (New York: AIP, 1984), pp. 157-161.
6. D. E. Holmgren, D. J. Walker, and S. E. Harris, "Emission at 1091 Å in Neutral Core-Excited Cs," in Laser Techniques in the Extreme Ultraviolet, S. E. Harris and T. B. Lucatorto, eds. (New York: AIP, 1984), pp. 496-501.
7. S. E. Harris, R. G. Caro, R. W. Falcone, D. E. Holmgren, J. E. Rothenberg, D. J. Walker, J. C. Wang, J. R. Willison, and J. F. Young, "Metastability in the XUV: Lasers and Spectroscopy," in Atomic Physics 9, R. S. Van Dyck, Jr. and E. N. Fortson, eds. (Singapore: World Scientific Publishing Co., 1985), pp. 462-479.
8. D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris, "Laser Spectroscopy of Na I Quartets," Phys. Rev. A 31, 677-683 (February 1985).
9. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "X-Ray Excitation of Energetic Metastable Levels in Atoms and Ions," Opt. News 11, 9-11 (March 1985).
10. A. J. Mendelsohn and S. E. Harris, "Proposal for an Extreme-Ultraviolet Selective Autoionization Laser in Zn III," Opt. Lett. 10, 128-130 (March 1985).
11. K. D. Pedrotti, A. J. Mendelsohn, R. W. Falcone, J. F. Young, and S. E. Harris, "XUV Emission Spectra of Core-Excited Levels in Sodium and Magnesium," J. Opt. Soc. Am. B (to be published).

12. S. E. Harris, J. F. Young, A. J. Mendelsohn, D. E. Holmgren, K. D. Pedrotti, and D. P. Dimiduk, "Quasi-Metastable Energy Levels and Applications," in Laser Spectroscopy VII, T. W. Hansch and Y. R. Shen, eds. (New York: Springer-Verlag, 1985), pp. 162-165.
13. S. E. Harris and R. G. Caro, "Shake-Up as a Mechanism for VUV Lasers," Opt. Lett. (to be published).
14. A. J. Mendelsohn, C. P. J. Barty, M. H. Sher, J. F. Young, S. E. Harris, "Emission Spectra of Quasi-Metastable Levels of Alkali Atoms," Phys. Rev. A (submitted for publication).

#### IV. LIST OF ORAL DISCLOSURES

1. "Metastable  $\text{Li}^+$  Levels Pumped by X-Rays From a Laser-Generated Plasma," Optical Society of America Meeting, Tucson, Arizona (October 1982).
2. "An Approach to X-Ray Lasers," University of Colorado, Boulder, Colorado (January 1983).
3. "Anti-Stokes Radiation for XUV Spectroscopy and Lasers," 13th Winter Colloquium on Quantum Electronics, Snowbird, Utah (January 1983).
4. "Anti-Stokes Radiation for VUV Spectroscopy and Lasers," Stanford University 1st Industrial Affiliates Symposium of the Department of Physics, Stanford, California (March 1983).
5. "XUV Lasers and High Resolution Radiation Sources," Symposium on Novel Sources of Electromagnetic Radiation, Germantown, Maryland (April 1983).
6. "Laser Induced Collisional Energy Transfer," American Physical Society DEAP Annual Meeting, Boulder, Colorado (May 1983).
7. "Anti-Stokes Scattering for Extreme Ultraviolet Lasers and Spectroscopy," Conference on Lasers and Electro-Optics (CLEO '83), Baltimore, Maryland (May 1983).
8. "Laser Spectroscopy in the Extreme Ultraviolet," Sixth International Conference on Laser Spectroscopy (SICOLS '83), Interlaken, Switzerland (June 1983).
9. "Laser Techniques for XUV Spectroscopy," Gordon Conference on Atomic Physics, New London, New Hampshire (July 1983).
10. "Soft X-Ray Pumping of Lithium," Gordon Research Conference on Atomic Physics, New London, New Hampshire (July 1983).
11. "Laser Spectroscopy in the Extreme Ultraviolet," Symposium on Atomic Spectroscopy (SAS-83), Berkeley, California (September 1983).
12. "Laser-Produced Plasmas: X-Ray 'Flashlamps' for the Excitation of Highly-Energetic States," Optical Society of America Meeting, New Orleans, Louisiana (October 1983).
13. "Spectroscopy of Core-Excited Levels for XUV Lasers," Sixth International Conference on Lasers and Applications (Lasers '83), San Francisco, California (December 1983).

14. "The Excitation of Metastable Extreme Ultraviolet Levels," OSA Second Topical Meeting on Laser Techniques in the Extreme Ultraviolet, Boulder, Colorado (March 1984).
15. "Quasi-Metastable Quartet Levels in Alkali-Like Atoms and Ions," OSA Second Topical Meeting on Laser Techniques in the Extreme Ultraviolet, Boulder, Colorado (March 1984).
16. "Grotrian Diagram of the Quartet System of Na I," OSA Second Topical Meeting on Laser Techniques in the Extreme Ultraviolet, Boulder, Colorado (March 1984).
17. "Metastability in the VUV: Lasers and Spectroscopy," Ninth International Conference on Atomic Physics, Seattle, Washington (July 1984).
18. "Metastability in the XUV - Lasers and Spectroscopy," Lawrence Livermore National Laboratory, Livermore, California (October 1984).
19. "Metastability in the XUV," Seventh International Conference on Lasers and Applications (Lasers '84), San Francisco, California (November 1984).
20. "Laser Produced X-Rays," Varian Associates, Palo Alto, California (January 1985).
21. "Quasi-Metastable Energy Levels and Applications Lasers," Seventh International Conference on Laser Spectroscopy (SEICOLS '85), Maui, Hawaii (June 1985).
22. "Ultraviolet Generation," Gordon Conference on Atomic Physics, Wolfeboro, New Hampshire (July 1985).
23. "Metastability in the Extreme Ultraviolet: Lasers and Spectroscopy," 1985 IBM Europe Institute Seminar on Laser Science, Lech, Austria (August 1985).

V. LIST OF PERSONNEL

Senior Personnel

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\* Ph.D. Degree awarded January 1985; "Excitation and Spectroscopy of Quartet Levels of Li, Na, and Cs Using a Pulsed Hollow-Cathode Discharge."

<sup>†</sup> Ph.D. Degree awarded June 1985; "Extreme Ultraviolet Emission Spectroscopy of Atoms and Laser Applications."

1473  
VI. ABSTRACTS OF PUBLICATIONS

4 LASER TECHNIQUES FOR EXTREME ULTRAVIOLET SPECTROSCOPY, *see page 1473*

by

S. E. Harris, J. F. Young, R. G. Caro, R. W. Falcone, D. E. Holmgren,  
D. J. Walker, J. C. Wang, Joshua E. Rothenberg, and J. R. Willison

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Stanford University  
Stanford, California 94035

ABSTRACT

In this paper we describe several techniques for using lasers to study core-excited energy levels in the spectral region between 10 eV and 100 eV. We are particularly interested in levels that are metastable against autoionization and, in some cases, against both autoionization and radiation.

ACTIVE MODE LOCKING OF A MICROWAVE-PUMPED XeCl Laser, *— 2nd page*

by

P. J. K. Wisoff and J. F. Young

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ABSTRACT

Acoustooptic mode locking of a long-pulse microwave-pumped XeCl laser has resulted in trains of ~ 320 ps long pulses having very high contrast ratios.

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MEASUREMENT OF LITHIUM AND SODIUM METASTABLE QUARTET ATOMS

IN A HOLLOW-CATHODE DISCHARGE; *→ next page*

by

D. E. Holmgren, R. W. Falcone, D. J. Walker, and S. E. Harris

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ABSTRACT

We report the measurement, in a pulsed hollow-cathode discharge, of metastable quartet atoms of Li and Na. By using a tunable probe laser, population densities of  $3 \times 10^{10}$  atoms  $\text{cm}^{-3}$  and  $10^{11}$  atoms  $\text{cm}^{-3}$  were measured in the  $\text{Li}(1s2s2p)^3P^0$  and  $\text{Na}(2p^33s3p)^3D_{7/2}$  levels, respectively. These levels are candidates for energy storage for extreme-ultraviolet lasers.

QUASI-METASTABLE QUARTET LEVELS IN ALKALILIKE ATOMS AND IONS, *→ kept page*

by

S. E. Harris, D. J. Walker, R. G. Caro, and A. J. Mendelsohn

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ABSTRACT

We describe the properties of a subclass of quartet levels of alkalilike atoms and ions that often retain metastability against autoionization and may have large radiative yields. Gain cross sections for XUV lasers with wavelengths between 20 and 100 nm are given.

✓ GROTRIAN DIAGRAM OF THE QUARTET SYSTEM OF Na I; → next page

by

D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris

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Stanford University  
Stanford, California 94035

ABSTRACT

Laser enhanced fluorescence experiments have been used in conjunction with Hartree-Fock calculations and emission studies to identify and locate Na I core-excited quartet levels. Twenty-eight Na emission lines are identified as transitions between these levels.

→ EMISSION AT 1091 Å IN NEUTRAL CORE-EXCITED Cs; → next page

by

D. E. Holmgren, D. J. Walker, and S. E. Harris

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Stanford, California 94305

#### ABSTRACT

Certain quartet levels in alkali-like systems retain metastability against autoionization while acquiring large radiative yields. This quasi-metastability occurs through selective coupling to non-autoionizing doublet levels by the spin-orbit interaction. An example of such a level is the  $5p^45d6s\ ^4P_{3/2}$  level of neutral Cs, which has a calculated branching ratio for radiation at 1091 Å of 43%. Experimentally, we find that this line has an emission intensity equal to 1/6 of that of the strongest ion line of  $\text{Cs}^+$ , and is a promising candidate for an extreme ultraviolet laser.

*Extreme Ultraviolet*

METASTABILITY IN THE XUV LASERS AND SPECTROSCOPY ; *→ next page*

by

S. E. Harris, R. G. Caro, R. W. Falcione, D. E. Holmgren, J. E. Rothenberg,  
D. J. Walker, J. C. Wang, J. R. Willison, and J. F. Young

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ABSTRACT

We describe the spectroscopy, methods for excitation, and applications of core-excited metastable atomic levels. Emphasis is on systems in alkali atoms and alkali-like ions which are metastable against autoionization and in some cases against radiation, and which allow lasing to a level in the valence structure of the atom.

✓ LASER SPECTROSCOPY OF Na I QUARTETS ; → next page

by

D. E. Holmgren, D. J. Walker, D. A. King, and S. E. Harris

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ABSTRACT

By using a pulsed hollow-cathode discharge, metastable atoms are produced in the  $(2p^3s3p)^4D_{7/2}$  and  $(2p^3s3p)^4S_{3/2}$  levels of Na I. A tunable laser is used to excite these atoms to levels in the  $2p^3s3d$  and  $2p^3s4s$  configurations, and thereby to establish a partial Grotrian diagram for the quartet system. Several possible Na I XUV laser systems are described.

X-RAY EXCITATION OF ENERGETIC METASTABLE LEVELS IN ATOMS AND IONS; *→ next page*

by

R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris

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ABSTRACT

This article describes an investigation of the excitation of energetic metastable levels suitable for use as storage levels in XUV laser systems. The production of large populations in excited levels with energies in the 200-100 eV region necessitates the development of excitation techniques characterized by high peak powers. The work reported here involves the use of an x-ray "flashlamp" as an excitation source. In addition, a secondary source of excitation has been developed in which the x-rays, emitted from the "flashlamp," are converted to a burst of high density energetic electrons. This device has been called a "photoionization electron source." Both of these excitation techniques have been shown to produce populations, in energetic (20-60 eV) metastable levels of atoms and ions, which are two to three orders of magnitude larger than have been achieved by alternative excitation methods.

PROPOSAL FOR AN EXTREME-ULTRAVIOLET SELECTIVE

AUTOIONIZATION LASER IN Zn III ; next page

by

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ABSTRACT

A system is proposed whereby Zn atoms that are photoionized by soft x-rays from a laser-produced plasma undergo selective super-Coster-Kronig decay leading to inversion and lasing on several XUV Zn III transitions. Calculations indicate that lasing will occur when a moderate-sized ( $\sim 10$  J)  $1.06 \mu\text{m}$  pump laser is used.

XUV EMISSION SPECTRA OF CORE-EXCITED LEVELS

IN SODIUM AND MAGNESIUM;

→ next page

by

K. D. Pedrotti, A. J. Mendelsohn, R. W. Falcone,  
J. F. Young, and S. E. Harris

Edward L. Ginzton Laboratory  
Stanford University  
Stanford, California 94305

ABSTRACT

Using a pulsed-hollow-cathode discharge we have observed the emission spectra of core-excited levels of Na I and Mg II. Line identifications and implications for XUV lasers are discussed.

↓  
QUASI-METASTABLE ENERGY LEVELS AND APPLICATIONS; *next page*

by

S. E. Harris, J. F. Young, A. J. Mendelsohn, D. E. Holmgren  
K. D. Pedrotti, and D. P. Dimiduk

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ABSTRACT

The paper summarizes the properties of certain radiating levels of the column I metals. XUV emission studies and an absorption experiment on the 1091 Å line of neutral Cs are described. A technique for correlating picosecond, broadband XUV pulses is discussed.

SHAKE-UP AS A MECHANISM FOR VUV LASERS; *next page*

by

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ABSTRACT

We show how electron shake-up, as it occurs during core photoionization, may produce population inversion in the vacuum ultraviolet. Calculations for Li show the possibility of lasers at 165.3 nm and 113.2 nm.

*and* EMISSION SPECTRA OF QUASI-METASTABLE LEVELS OF ALKALI ATOMS . 

by

A. J. Mendelsohn, C. P. J. Barty, M. H. Sher,  
J. F. Young, and S. E. Harris

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Stanford, California 94305

ABSTRACT

The paper describes the properties of a subclass of quartet levels of the alkali atoms which often retain metastability against autoionization and may have large radiative yields. Using high pulsed power microwaves, we obtain emission spectra of Na, K, Rb, and Cs. In each case, the neutral emission spectrum is dominated by emission from these levels.